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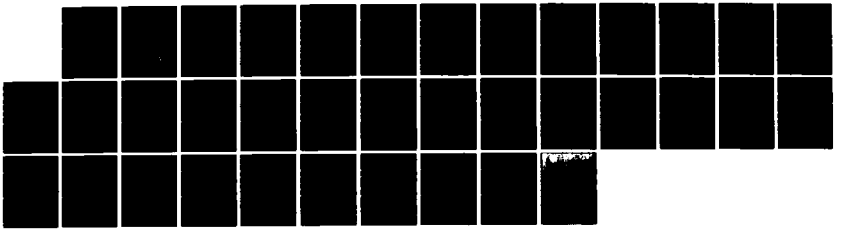

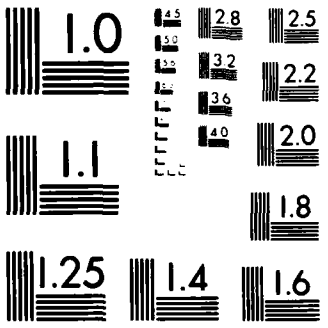

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Raman Spectroscopic Study of Molecular Orientation in AgTCNQ Thin Films

by

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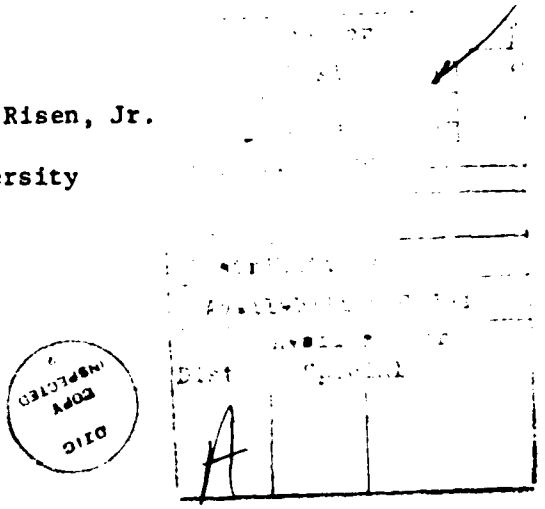

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Abstract

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Introduction

The synthesis, structure and physical properties of TCNQ charge-transfer salts have been the subject of extensive investigations over the past two decades. It is known from X-ray diffraction studies that the common structural characteristic of these materials is face to face stacking of the TCNQ moities to form pseudo-one dimensional columns, which may be along one crystallographic axis (1) or tilted relative to a crystallographic axis (2). This directionality of packing of the TCNQ units results in anisotropic extensive physical properties, especially their electrical conductivity and magnetic and optical properties. Such details of the mode of packing as the formation of segregated (3) as opposed to mixed stacks (4) of donor and TCNQ molecules, and the degree of charge transfer from donor to TCNQ (5), determine the absolute values of the extensive properties in different directions without affecting the anisotropic character of the solid state TCNQ salts.

The reported X-ray structural results on TCNQ-compounds were obtained on single crystals of salts prepared by solution methods. We have shown (6,7) that in addition to being preparable by these solution methods of Melby et al. (8), CuTCNQ and AgTCNQ can be prepared as thin films on a variety of substrates by a thermal method. In this method, alternating layers of the metal and TCNQ, of the appropriate thicknesses, are vapor-deposited on a substrate and then heated at ca 100°C to cause a direct reaction of metal with TCNQ. The MTCNQ-films obtained that way have the same

spectral features as those prepared by the solution methods, and we have found that they exhibit interesting resonance Raman (9) and photo-transformation phenomena (7).

The crystal structures of CuTCNQ and AgTCNQ are not known, but it is likely that crystals of these salts are characterized by stacks of $TCNQ^-$ units, as are the other TCNQ-compounds prepared by similar solution procedures. Since the thin films can have important properties due to their form, it is of interest to investigate whether or not such stacking occurs in them as well or whether the $TCNQ^-$ units in thermally prepared thin films are disordered in some way. If preferred orientation of $TCNQ^-$ units occurs in thin films, it is expected that they would exhibit interesting directional semiconducting properties. If there is no such order, a new basis for understanding their electronic and optical properties will need to be developed.

Vibrational spectrsocopy can be employed to measure the existence and the extent of molecular orientation in a variety of materials. For example Raman depolarization and infrared dichroism measurements have been used to determine the degree of molecular orientation in polymeric fibers (10-12), and to study chain conformations in proteins (13). Recently, we have used polarized Raman scattering to probe the extent of molecular orientation in glass systems. Thus, we showed that $(PO_3)_n^{-n}$ metaphosphate chains are preferentially oriented with the axis of $(NaPO_3)_x$ glass fibers drawn from the melt (14), and that boroxol rings are preferentially oriented in response to an applied stress in blown films of B_2O_3 glass (15).

In this study polarized Raman spectroscopy has been employed to explore the existence of preferred orientation of the $TCNQ^-$ units in AgTCNQ thin films prepared by the thermal method. The experimental results for Raman intensities as a function of sample orientation are compared with those predicted for films with $TCNQ^-$ units with various postulated types of orientation relative to the substrate and to each other.

Experimental

The polarized Raman scattering measurements were performed on AgTCNQ thin films since they are of better optical quality than CuTCNQ thin films and exhibit analogous phenomena. The preparation of thin films has been described previously (6,7). Briefly, films of ca 2000 Å thickness were prepared on three substrates that have different electrical conductivities. A metal, aluminum film (ca 5 μm thick vapor deposited on a fused silica plate), an insulator (a flat fused silica plate),and a semiconductor, Si(100) wafer, were the substrates. To provide a basis for comparison, polarized Raman spectra also were measured on a CH_3CN solution of AgTCNQ, in which the $TCNQ^-$ ions are randomly oriented. The AgTCNQ powder from which the dilute solution was made was prepared by the metathetical reaction of $AgNO_3$ and LiTCNQ (8).

Raman spectra were obtained on a Jarrell-Ash 25-300 laser Raman spectrometer using the 632.8 nm line of a Spectra Physics 125A He-Ne laser. A 90° scattering geometry was used, with Z defined as the direction of propagation of the incident light and X as that of the scattered light. The plane of polarization of the incident light was controlled by a half-wave plate, coated for optimal performance at 632.8 nm. The polarization of the scattered light was determined using a Polaroid sheet analyzer. To compensate for the difference in response of the gratings and other optical components of the spectrometer for different polarizations of the light entering the spectrometer, a scrambler was placed before the entrance slit.

The convention used to describe the polarized Raman spectra, i(kl)j, is that of Porto et al. (16). The symbols outside the parentheses to the left and the right are the propagation directions of the incident and scattered light respectively. The symbols inside the parentheses, left to right, are the polarization directions of the incident and scattered light. The Raman depolarization data were obtained for the totally symmetric (Ag) mode of AgTCNQ at ca 1605 cm^{-1}, which is the strongest band of the spectrum for this particular excitation wavelength. The

optical arrangements and spectrometer response were calibrated by measuring the depolarization ratios of CCl_4. The depolarization ratios obtained are $\rho = 0.010 \pm 0.001$ for the 459 cm^{-1} band, and $\rho = 0.75 \pm 0.01$ for the 314 and 218 cm^{-1} band, in excellent agreement with the established values (17).

Theory

For randomly oriented molecules the power of the Raman scattered radiation is determined by the space-averaged value of the square of the induced dipole moment. Thus, the average of the squares of the derived polarizability tensor components, over all orientations of the molecular-fixed axes (x,y,z) relative to the laboratory-fixed axes (X,Y,Z) are required. Then, the Raman intensities are functions of two quantities, which are invariant under any transformation of coordinate system. These quantities are the mean polarizability $\bar{\alpha}$ and the anisotropy γ, functions of the derived molecular polarizability tensor elements, α_{pq}, where

$$\alpha_{pq} = \left(\frac{\partial^2 \alpha}{\partial p \partial q} \right)_o$$

and

$$\bar{\alpha} = \frac{1}{3}(\alpha_{xx} + \alpha_{yy} + \alpha_{zz}) \qquad (1)$$

$$\gamma^2 = \frac{1}{2}[(\alpha_{xx}-\alpha_{yy})^2+(\alpha_{yy}-\alpha_{zz})^2+(\alpha_{zz}-\alpha_{xx})^2+6(\alpha_{xy}^2+\alpha_{yz}^2+\alpha_{zx}^2)] \qquad (2)$$

The space averages of the squares of the scattering tensor components, in laboratory coordinates, are calculated to be (18,19):

$$\langle\alpha_{XX}^2\rangle = \langle\alpha_{YY}^2\rangle = \langle\alpha_{ZZ}^2\rangle = \frac{45(\bar{\alpha})^2+4\gamma^2}{45} \qquad (3)$$

$$\langle\alpha_{XY}^2\rangle = \langle\alpha_{YZ}^2\rangle = \langle\alpha_{XZ}^2\rangle = \frac{\gamma^2}{15} \qquad (4)$$

In the case of partial or complete molecular orientation eqn (3) and (4) do not hold and the Raman band intensities depend explicitly on the experimental geometry, the position of the molecular coordinates relative to the laboratory coordinates.

For AgTCNQ thin films we postulate that if there is a preferred orientation of the $TCNQ^-$ units relative to the substrate, it is as shown schematically in Figure 1(a). Specifically, this postulate is that each $TCNQ^-$ unit is oriented in such a way that the two CN groups at the same end of the molecule are pointing towards the substrate and that the molecular plane makes an angle θ with the plane of the substrate.

In Figure 1(a), x is defined as the long molecular axis, while the short molecular axis y is parallel to the substrate, under the assumptions made here. In Figure 1(b) the molecular axes (x,y,z) are shown relative to the substrate axes (A,B,C), with C defined as being perpendicular to the substrate plane. To specify any coordinate system relative to another three Euler angles are required, and in this paper the Euler angle definitions of Wilson-Decius and Cross are adopted (18). Two of the Euler angles are designated as θ and ϕ in Figure 1(b). The third angle, ψ, is defined as the angle that the y axis makes with the intersection of the (AB) and (xy) planes. Since y is here parallel to the substrate, ψ is zero in Figure 1(b). So the two angles, θ (fixed by the postulate) and ϕ, define completely the (x,y,z) coordinate system relative to the substrate (A,B,C) coordinates. While this postulate is that all $TCNQ^-$ units are oriented in the same way relative the the substrate, there is no obvious reason to believe that all $TCNQ^-$ units sampled by the source beam are parallel to each other, since relatively large film areas are sampled. It is more likely that the way the film differs from a crystal is that there are domains such that the $TCNQ^-$ units in one are parallel to each other but not parallel to units of other domains even though the orientation of each unit relative to the substrate is the same. This is equivalent to saying that θ is fixed but $0 \leq \phi \leq 2\pi$.

In the Raman scattering experiments we can control only the orientation of the substrate relative to the laboratory coordinates in addition to the control of the

polarization. For these measurements the substrate was oriented with its C axis in the (XZ) plane, Figure 1(c). Measurements in two other orientations, where C was in the (XY) or (YZ) planes, also were tried but the measured intensities were too weak to give reliable intensity ratios. So, we consider here only the case where C is in the (XZ) plane. The angle β is experimentally determined and takes values from zero to 90°. The equivalent to angle ϕ (of Figure 1(b)) is zero in Figure 1(c). The third angle, ψ, specifies the variation of A and B axes, within the substrate plane, and is arbitrary. To simplify the calculations we set $\psi = 0$ (Figure 1(c)), but even if ψ $(0 \leq \psi \leq 2\pi)$ is included and then averaged over all possible values the expressions obtained for the averages of the squares of the tensor elements (in X,Y,Z coordinates) are the same as are obtained with $\psi = 0$.

Since the intensity of Raman scattered light is directly proportional to the space-averaged value of the square of the relevant α_{PQ} element, the derived polarizability tensor elements in (X,Y,Z) coordinates, in terms of derived molecular polarizability elements α_{pq}, are needed. Here P is the direction of polarization of the incident light and Q is that of the scattered light. To obtain these expressions needed, the components of the polarizability tensor in substrate coordinates, α_{FG}, are first expressed in terms of α_{pq} by:

$$\alpha_{FG} = T_1 \alpha_{pq} \tilde{T}_1 \ , \qquad (5)$$

where T_1 is the transformation matrix relating the (x,y,z) and (A,B,C) axes systems: $(A,B,C) = T_1(x,y,z)$ and $\tilde{T}_1$ is its transpose. The elements of matrix T_1 are the direction cosines, that is the cosines axis A makes with axes x, y and z, and so on. The form of matrix T_1 is:

$$T_1 = \begin{pmatrix} \cos\theta\cos\phi & -\sin\phi & \sin\theta\cos\phi \\ \cos\theta\sin\phi & \cos\phi & \sin\theta\sin\phi \\ -\sin\theta & 0 & \cos\theta \end{pmatrix} \qquad (6)$$

The α_{PQ} elements are now expressed in terms of α_{FG} by:

$$\alpha_{PQ} = T_2 \, \alpha_{FG} \, \tilde{T}_2 \tag{7}$$

Here, T_2 is the transformation matrix relating the (X,Y,Z) and (A,B,C) axes systems: $(X,Y,Z) = T_2(A,B,C)$ and is given by:

$$T_2 = \begin{pmatrix} \cos\beta & 0 & \sin\beta \\ 0 & 1 & 0 \\ -\sin\beta & 0 & \cos\beta \end{pmatrix} \tag{8}$$

Combining eqn (5) and (7) results in the following expression for α_{PQ}:

$$\alpha_{PQ} = T_2 T_1 \alpha_{pq} \tilde{T}_1 \tilde{T}_2 = (T_2 T_1) \alpha_{pq} (\widetilde{T_2 T_1}) \tag{9}$$

In a tensor form α_{PQ} is written as:

$$\alpha_{PQ} = \begin{pmatrix} \alpha_{XX} & \alpha_{XY} & \alpha_{XZ} \\ \alpha_{YX} & \alpha_{YY} & \alpha_{YZ} \\ \alpha_{ZX} & \alpha_{ZY} & \alpha_{ZZ} \end{pmatrix} \tag{10}$$

If we assume that $TCNQ^-$ retains the molecular symmetry (D_{2h}) of the neutral species, then the polarizability tensor in molecular coordinates and Ag vibrational modes is given as follows (20,21):

$$\alpha_{pq} = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix} \tag{11}$$

For the 90° scattering geometry four Raman configurations are possible: Z(YY)X, Z(YZ)X, Z(XZ)X and Z(XY)X, so the tensor elements α_{YY}, α_{YZ}, α_{XZ} and α_{XY} are needed to compute Raman scattering intensities. By employing eqn (9) and (11) and the expressions for T_1 and T_2 these four elements are calculated. When squared and averaged over all possible values of ϕ, the results are:

$$\langle \alpha_{YY}^2 \rangle = \frac{1}{8}[(a\cos^2\theta + c\sin^2\theta + b)^2 + (\sqrt{2}\, a\cos^2\theta + \sqrt{2}\, c\sin^2\theta)^2 + 2b^2] \quad (12)$$

$$\langle \alpha_{YZ}^2 \rangle = \frac{1}{8}(a\cos^2\theta + c\sin^2\theta - b)^2 \sin^2\beta + \frac{1}{2}(a-c)^2(\cos^2\theta \sin^2\theta)\cos^2\beta \quad (13)$$

$$\langle \alpha_{XZ}^2 \rangle = \frac{1}{8}[4(a-c)^2(\sin^2\theta - \cos^2\theta)^2 + 4(a\sin^2\theta + c\cos^2\theta - b)^2 -$$

$$-(a\cos^2\theta + c\sin^2\theta - b)^2]\cos^2\beta \sin^2\beta + \frac{1}{2}(a-c)^2(\cos^2\theta \sin^2\theta)(\sin^2\beta - \cos^2\beta)^2 \quad (14)$$

$$\langle \alpha_{XY}^2 \rangle = \frac{1}{8}(a\cos^2\theta + c\sin^2\theta - b)^2\cos^2\beta + \frac{1}{2}(a-c)^2(\cos^2\theta \sin^2\theta)\sin^2\beta \quad (15)$$

Thus the various $\langle \alpha_{PQ}^2 \rangle$ are now expressed in terms of molecular polarizability elements and trigonometric functions of angles θ and β.

Results and Discussion

AgTCNQ Solution Spectra

Representative polarized Raman spectra of AgTCNQ in CH_3CN solution are shown in Figure 2, for the four different combinations of the polarization of the incident and scattered light. The measured intensities are normalized relative to the intensity of the Z(YY)X orientation, so that three intensity ratios are obtained experimentally. The ratios reported here are the average of at least three sets of experiments; $\langle \alpha_{YZ}^2 \rangle / \langle \alpha_{YY}^2 \rangle = 0.33 \pm 0.01$, $\langle \alpha_{XZ}^2 \rangle / \langle \alpha_{YY}^2 \rangle = 0.34 \pm 0.01$ and $\langle \alpha_{XY}^2 \rangle / \langle \alpha_{YY}^2 \rangle = 0.33 \pm 0.01$. These results are in excellent agreement with the theoretically predicted relative intensities for complete randomness, since they are equal, within experimental error, as predicted by eqns. (4). The three measured ratios give an average of 0.33 ± 0.01 for this Ag mode of $TCNQ^-$ in solution.

For this particular vibrational mode, which is an in-plane, totally symmetric mode (22), it is reasonable to set $c = \alpha_{zz} = 0$. Using $\alpha_{xx} = a$ and $\alpha_{yy} = b$, $\alpha_{ij} = 0$ for $i \neq j$ and combining eqn. (1), (2), (3) and (4) the calculated values of R are:

$$R = \frac{\langle \alpha^2_{YZ} \rangle}{\langle \alpha^2_{YY} \rangle} = \frac{\langle \alpha^2_{XZ} \rangle}{\langle \alpha^2_{YY} \rangle} = \frac{\langle \alpha^2_{XY} \rangle}{\langle \alpha^2_{YY} \rangle} = \frac{a^2+b^2-ab}{3a^2+3b^2+2ab} \qquad (16)$$

The ratio R approaches the value $R = \frac{1}{3}$ only when $\frac{a}{b}$ (or $\frac{b}{a}$) is very large and becomes equal to $\frac{1}{3}$ when $a \neq 0$, $b=0$ or $b \neq 0$, $a=0$.

Thus, the solution results suggest that one of the non-zero diagonal elements of the α_{pq} tensor is very large relative to the others. The 632.8-nm line used for Raman excitation is within the red band system of $TCNQ^-$ (23,24), so the 1605 cm^{-1} band is resonantly enhanced. The red band system involves the $^2B_{2g} \rightarrow {}^2B^{(1)}_{1u}$ electronic transition, which is x-polarized (23), so it is reasonable to assume that, due to resonance enhancement, the $\alpha_{xx} = a$ element will be much larger than $\alpha_{yy} = b$. Thus, the Raman depolarization data obtained from the AgTCNQ-CH_3CN solution spectra show the kind of relative intensities expected for randomly oriented $TCNQ^-$ units, with $a >> b$.

AgTCNQ Thin Film Spectra.

Representative polarized Raman spectra of AgTCNQ thin films on Al substrate are shown in Figure 3, for different values of angle β. Comparison with the solution spectra of Figure 2, shows that there is a remarkable difference in relative intensities and a strong dependence on angle β. This observation shows that some orientation of the $TCNQ^-$ units relative to the substrate must exist. The experimental relative intensities for different substrates and orientation angle β are given in Table 1.

The comparison of the experimental relative intensities with the ones predicted on the basis of the postulates developed above can now be used to determine the orientation in AgTCNQ thin films. Using the relations derived in the previous section, with $c = 0$ and $a >> b$, the general expressions given in eqns (12)-(15) can now be simplified to yield:

$$\langle a_{YY}^2 \rangle = \frac{3}{8} a^2 \cos^4\theta \tag{17}$$

$$\langle a_{YZ}^2 \rangle = \frac{1}{8} a^2 \cos^4\theta \sin^2\beta + \frac{1}{2} a^2(\cos^2\theta \sin^2\theta)\cos^2\beta \tag{18}$$

$$\langle a_{XZ}^2 \rangle = \frac{1}{8} (8 \sin^4\theta + 3 \cos^4\theta - 8 \sin^2\theta \cos^2\theta)a^2(\cos^2\beta \sin^2\beta) +$$

$$+ \frac{1}{2} a^2(\cos^2\theta \sin^2\theta)(\sin^2\beta - \cos^2\beta)^2 \tag{19}$$

$$\langle a_{XY}^2 \rangle = \frac{1}{8} a^2 \cos^4\theta \cos^2\beta + \frac{1}{2} a^2(\cos^2\theta \sin^2\theta)\sin^2\beta \tag{20}$$

Since only $\langle a_{YY}^2 \rangle$ is independent of β, the relative intensities with respect to that orientation are obtained.

$$\langle a_{YZ}^2 \rangle / \langle a_{YY}^2 \rangle = \frac{1}{3} \sin^2\beta + \frac{4}{3} \tan^2\theta \cos^2\beta \tag{21}$$

$$\langle a_{XZ}^2 \rangle / \langle a_{YY}^2 \rangle = \frac{1}{3} (8 \tan^4\theta - 8 \tan^2\theta + 3)\cos^2\beta \sin^2\beta + \frac{4}{3} \tan^2\theta(\sin^2\beta - \cos^2\beta)^2 \tag{22}$$

$$\langle a_{XY}^2 \rangle / \langle a_{YY}^2 \rangle = \frac{1}{3} \cos^2\beta + \frac{4}{3} \tan^2\theta \sin^2\beta \tag{23}$$

The derived expressions for the theoretical relative intensities are functions of β and θ only. Since θ is fixed, but unspecified by the model, and β is the experimental angle, these relative intensity functions (eqns (21)-(23)) can be plotted versus β, for different values of θ. The calculated curves are shown in Figures (4), (5) and (6), as solid lines. By calculating the first and second derivative (with respect to β) on the right hand side of eqns (21)-(23) it is easy to show that the $\langle a_{YZ}^2 \rangle / \langle a_{YY}^2 \rangle$ and $\langle a_{XY}^2 \rangle / \langle a_{YY}^2 \rangle$ functions have an inflection point at $\beta = 45°$, for $0 \leq \beta \leq 90°$. Also the function $\langle a_{XZ}^2 \rangle / \langle a_{YY}^2 \rangle$ has inflection points at $\beta = 22.5°$ and $\beta = 67.5°$, while the specific value of angle θ determines whether it shows a maximum or minimum value. Thus for $\theta < 19.88°$ or $\theta > 59.44°$ it shows a maximum at $\beta = 45°$, while for $19.88° < \theta < 59.44°$ the function shows a minimum at $\beta = 45°$.

Finally for $\theta = 19.88°$ or $\theta = 59.44°$ the $\langle\alpha^2_{XZ}\rangle/\langle\alpha^2_{YY}\rangle$ function does not depend on β and it is a constant determined by θ.

In Figures (4), (5) and (6) we also have plotted the experimental relative intensities for different orientation angle β and various substrates. It is shown that for all three relative intensities there is good agreement between experimental values and calculated curves, especially in the observed trends. The experimental values of $\langle\alpha^2_{XZ}\rangle/\langle\alpha^2_{YY}\rangle$ relative intensity show well defined minima at $\beta = 45°$, for all three substrates, in excellent agreement with the predicted curves. These experimental values indicate that an angle θ in the 45° to 52.5° range is expected. The other two experimental relative intensities, $\langle\alpha^2_{YZ}\rangle/\langle\alpha^2_{YY}\rangle$ and $\langle\alpha^2_{XY}\rangle/\langle\alpha^2_{YY}\rangle$, show also the predicted trends, and an orientation angle θ in the 40° to 45° range. Thus, the main experimental result of this investigation is that the predicted relative intensity patterns are observed in the $\theta = 40°$ to 52.5° range.

In Raman depolarization studies of this kind there are several experimental errors and optical effects that can affect, to some extent, the experimental observations. For example, for ratios involving weak bands, noise is one source of error. Low scanning speed and high time constant were used to reduce this source of potential error. Microdefects of the thin films can also cause the incident light to lose its direction of propagation and its initial polarization. Experimental errors, and thus deviations from the predicted results, are expected to be greater for $\beta = 0°$ and 90°, due to the fact that the thin films are substrate-supported and so the interference of the substrate with the incident or scattered light will be a maximum for $\beta = 0°$ and 90° respectively.

The optical effects that are important to consider in these measurements, are refraction and polarization-selective reflection. Since AgTCNQ is an anisotropic material, birefringence or double refraction is expected even at normal incidence.

Thus, for each orientation angle the beam will be refracted and split into two beams with polarization orientations that are mutually orthogonal. The optical indicatrix (index ellipsoid) for these films is not known, so it is impossible to estimate the incident beam deviation or its polarization change, or thus their effect on experimental intensity ratios. The reflected or transmitted light is polarization-selective. For a homogeneous and isotropic medium the transmitted light polarized in the plane of incidence (XZ) is of higher intensity than the transmitted light polarized perpendicularly, (YY), to the plane of incidence (25). If we assume that a fraction of the Raman scattering is excited by the transmitted light, then higher intensity light is available for the (XZ) than the (YY) scattering configuration. This may be the reason that the $\langle\alpha^2_{XZ}\rangle/\langle\alpha^2_{YY}\rangle$ relative intensities are displaced to higher θ values (40°-52.5°) compared to θ = 40°-45° obtained from the other two relative intensities. AgTCNQ thin films are highly absorbing (with 632.8 nm excitation), so it is expected that most of the measured Raman scattering is generated at the surface or within a small thickness of the film. In this case it is expected that the impact of different optical phenomena on our measurements will be smaller than for transparent materials. In any event, regardless the effect of the optical phenomena on absolute Raman intensities, the effect on the relative intensities is expected to be small, so the observed patterns are the true ones.

In conclusion the data show that AgTCNQ thin films are characterized by preferred orientation of the $TCNQ^-$ units relative to the substrate, and that agreement of the experimental results with the calculated relative intensities is obtained with $\theta \simeq 45°$. The presence of this (θ) ordering and the lack of other orientation in the sampled region is taken to indicate that there are many relatively small domains, within each of which $\theta \simeq 45°$, which are not ordered with respect to each other.

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Table 1

Experimental Relative Intensities for Different Substrates and Angle β

	$\langle\alpha^2_{YZ}\rangle/\langle\alpha^2_{YY}\rangle$			$\langle\alpha^2_{XY}\rangle/\langle\alpha^2_{YY}\rangle$		
β	Si	SiO_2	Al	Si	SiO_2	Al
90	0.46	0.42	0.42	1.86	2.15	2.04
75	0.46	0.45	0.44	0.81	1.09	0.97
60	0.46	0.48	0.51	0.61	0.81	0.71
45	0.56	0.56	0.61	0.47	0.80	0.56
30	0.58	0.83	0.80	0.49	0.53	0.50
15	1.01	0.91	1.00	0.45	0.51	0.49
0	1.62	1.08	1.10	0.48	0.47	0.38

$\langle\alpha^2_{XZ}\rangle/\langle\alpha^2_{YY}\rangle$

β	Si	SiO_2	Al
90	3.18	3.05	3.69
75	1.40	2.05	1.83
60	1.02	1.45	1.38
45	0 77	1.12	1.01
30	0.88	1.38	1.28
15	1.67	1.66	1.86
0	2.59	2.25	1.79

Figure Captions

Figure 1. Definition of the molecular (x,y,z), substrate (A,B,C) and laboratory (X,Y,Z) coordinate system.

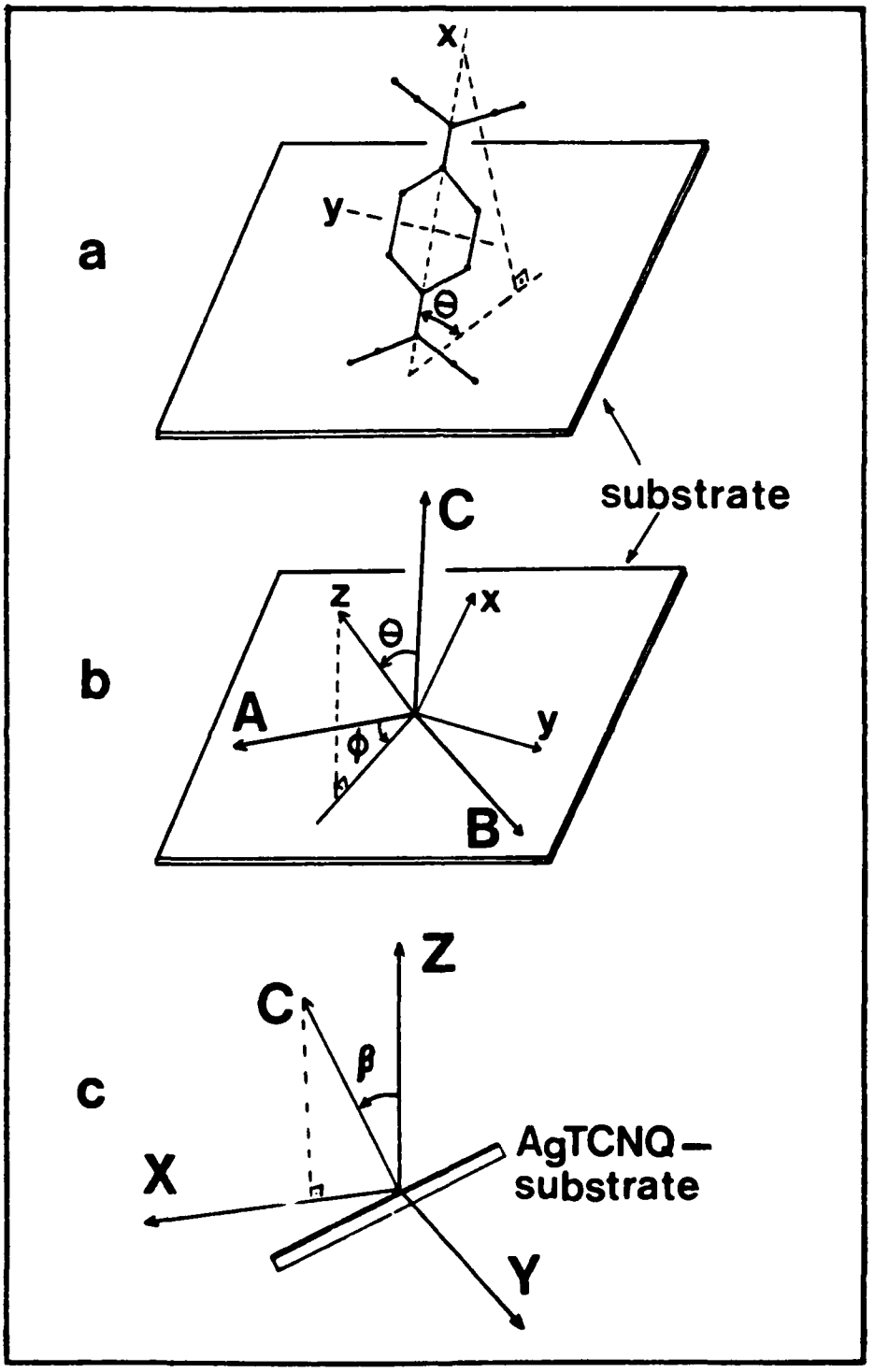
Figure 2. Polarized Raman spectra of AgTCNQ-CH_3CN solution.

Figure 3. Polarized Raman spectra of AgTCNQ thin films, at different angles β.

Figure 4. $\langle\alpha^2_{YZ}\rangle/\langle\alpha^2_{YY}\rangle$ relative intensity. Solid lines represent curves calculated from eqn (21), for different θ values. Points represent experimental values for thin films on various substrates: O Si(100), Δ SiO_2 and □Al.

Figure 5. $\langle\alpha^2_{XZ}\rangle/\langle\alpha^2_{YY}\rangle$ relative intensity. Solid lines represent curves calculated from eqn (22), for different θ values. Points represent experimental values for thin films on various substrates: O Si(100), Δ SiO_2 and □ Al.

Figure 6. $\langle\alpha^2_{XY}\rangle/\langle\alpha^2_{YY}\rangle$ relative intensity. Solid lines represent curves calculated from eqn (23), for different θ values. Points represent experimental values for thin films on various substrates: O Si(100), Δ SiO_2, and □ Al.


a
x
y
θ
substrate
b
C
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y
φ
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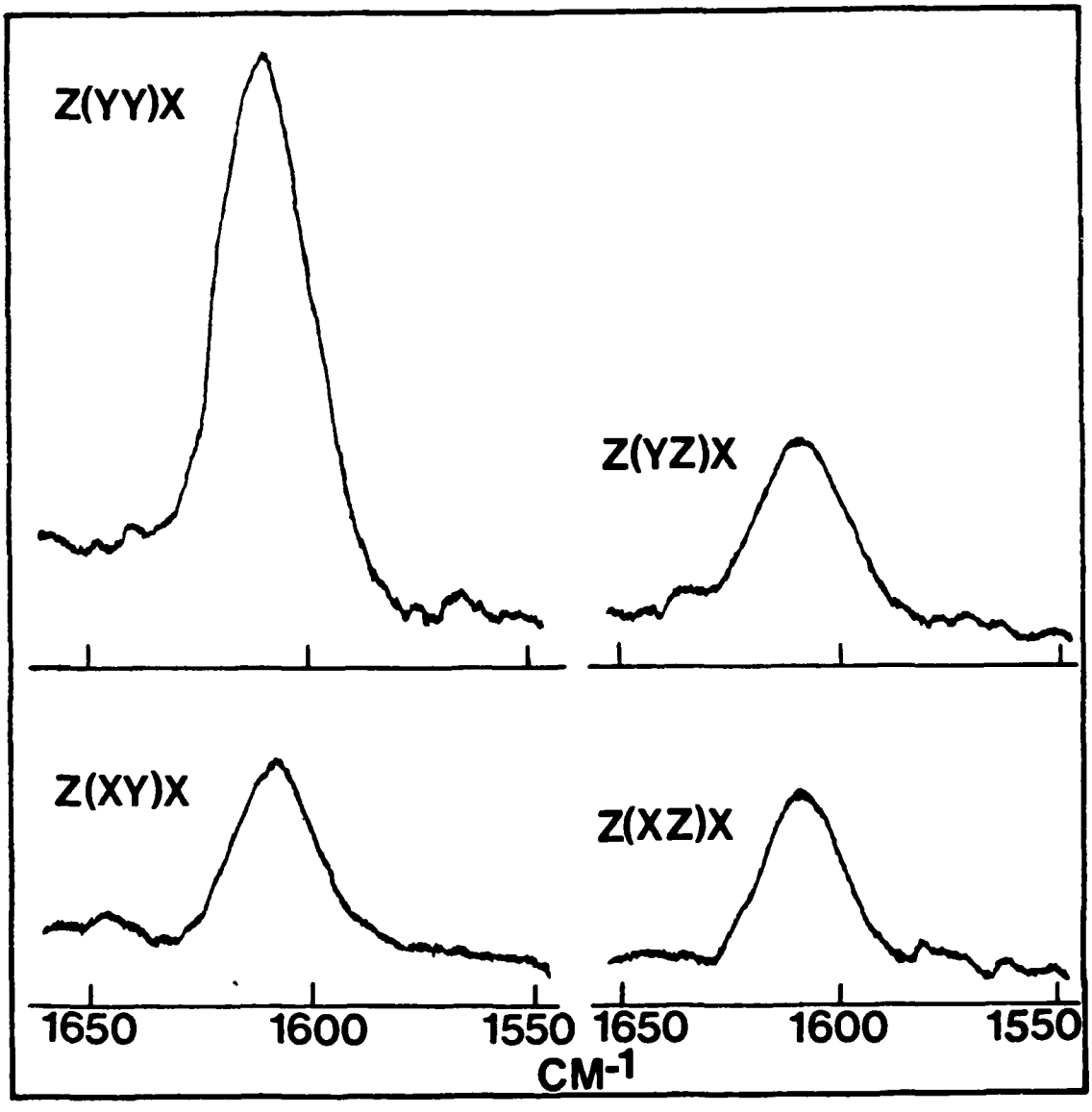
substrate
Y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Z(XZ)X
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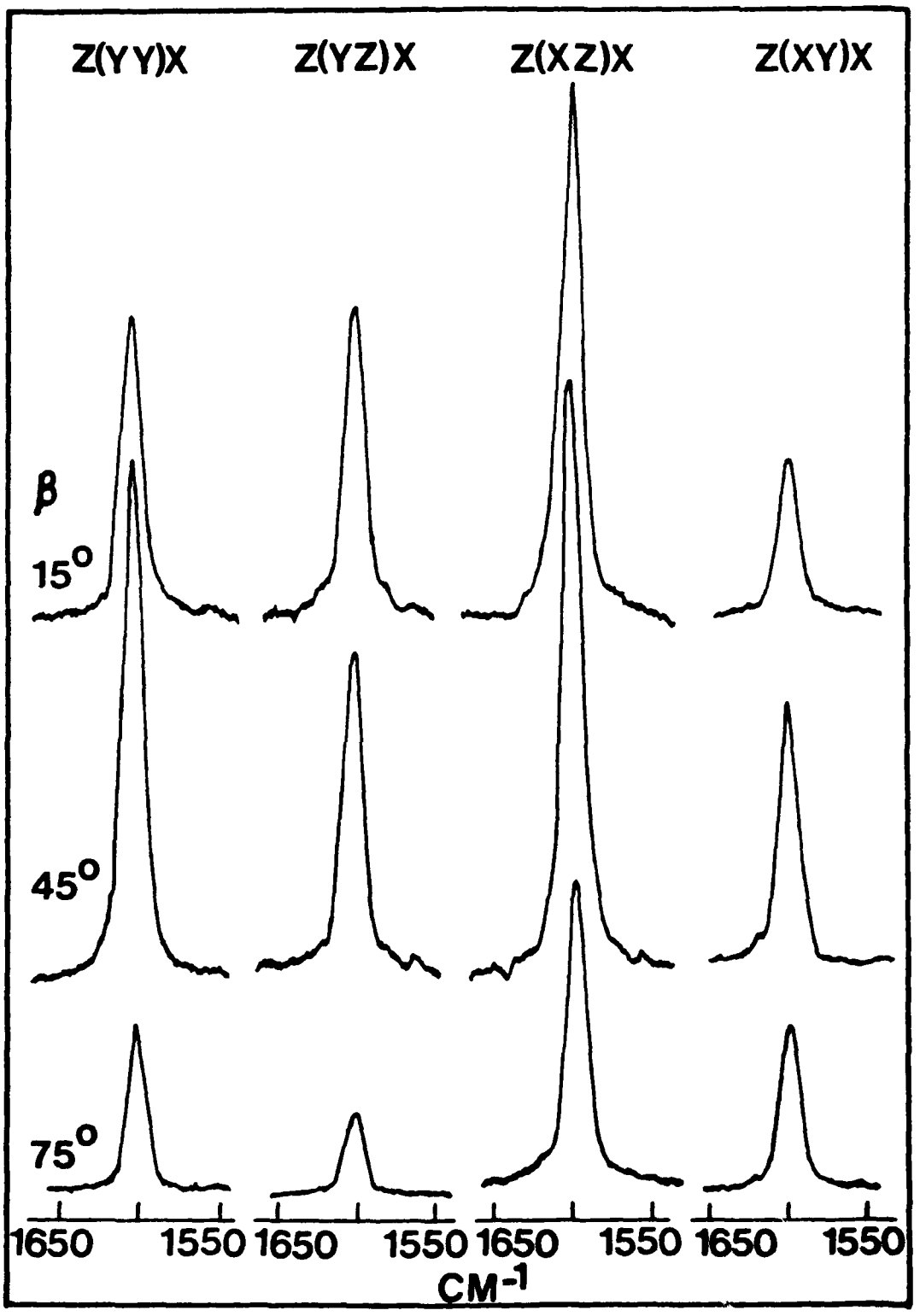
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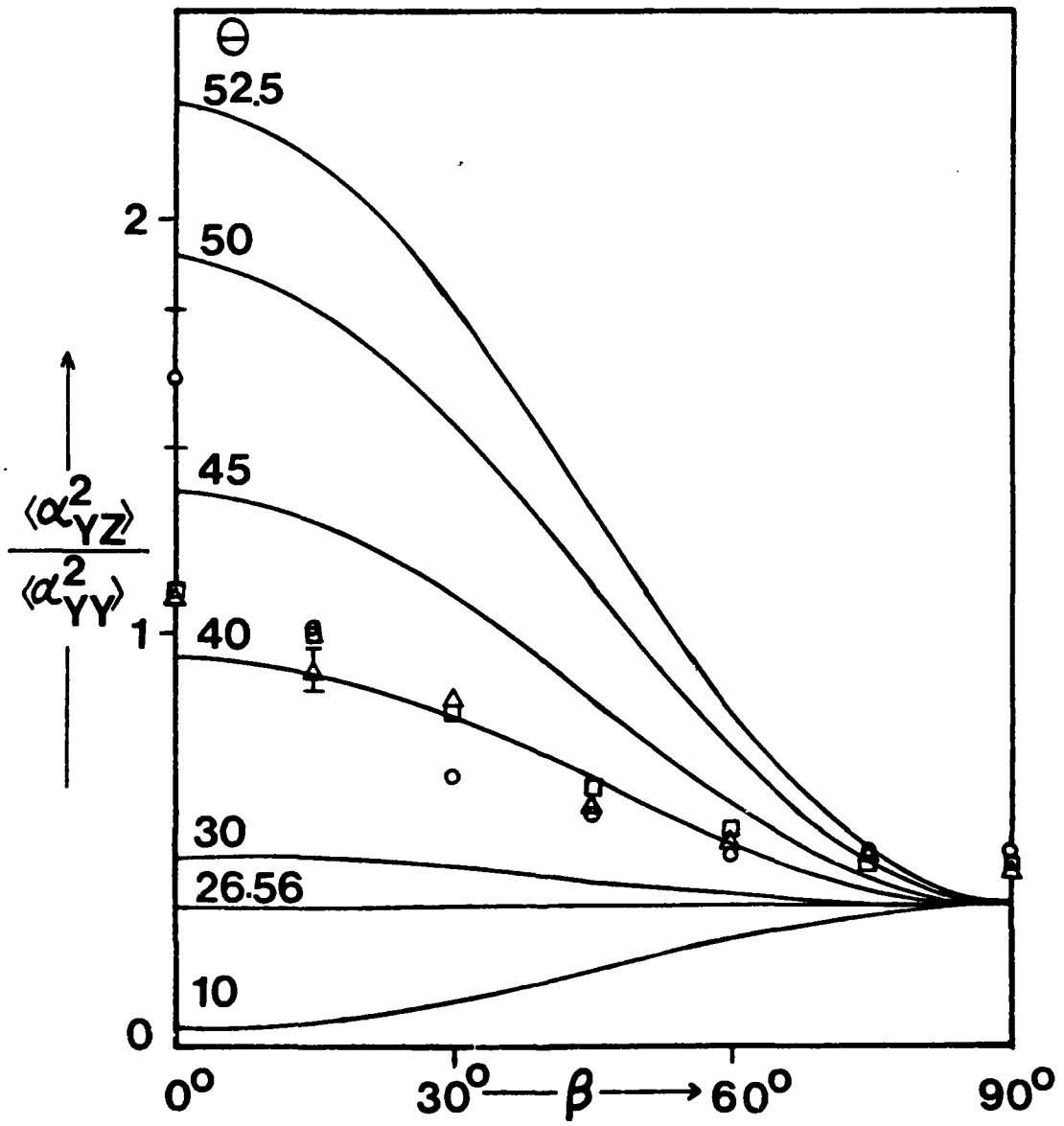
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β
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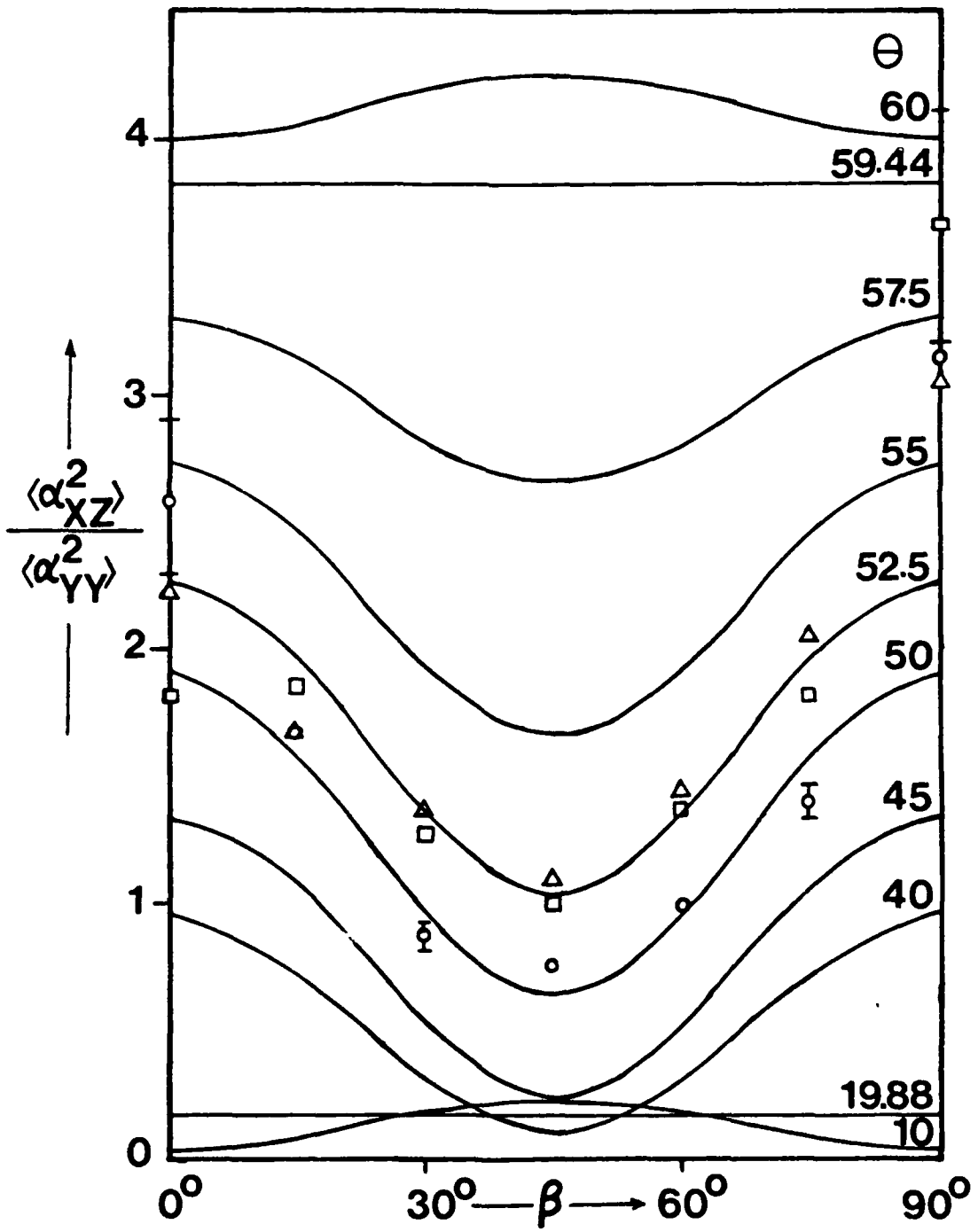
90°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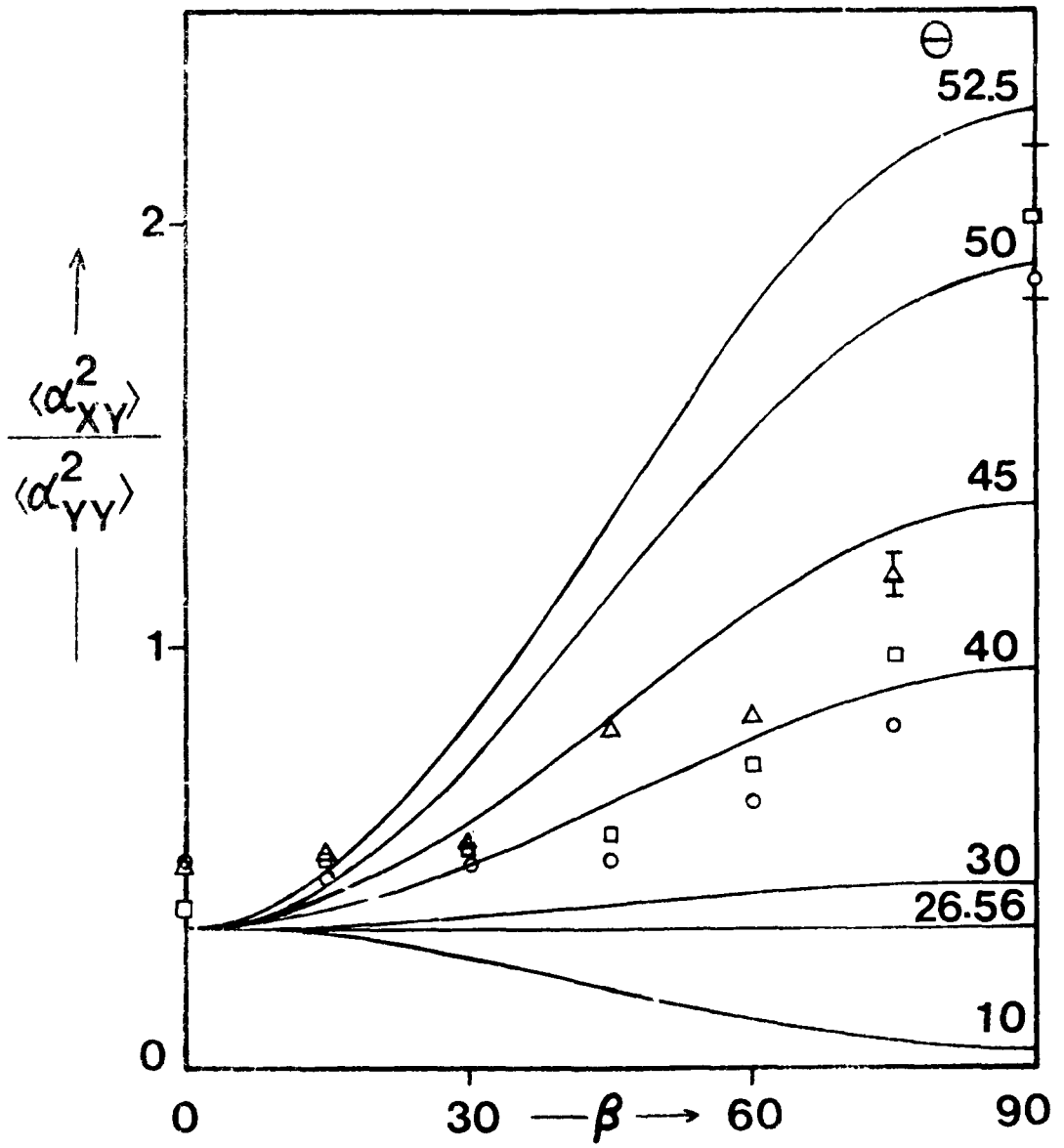
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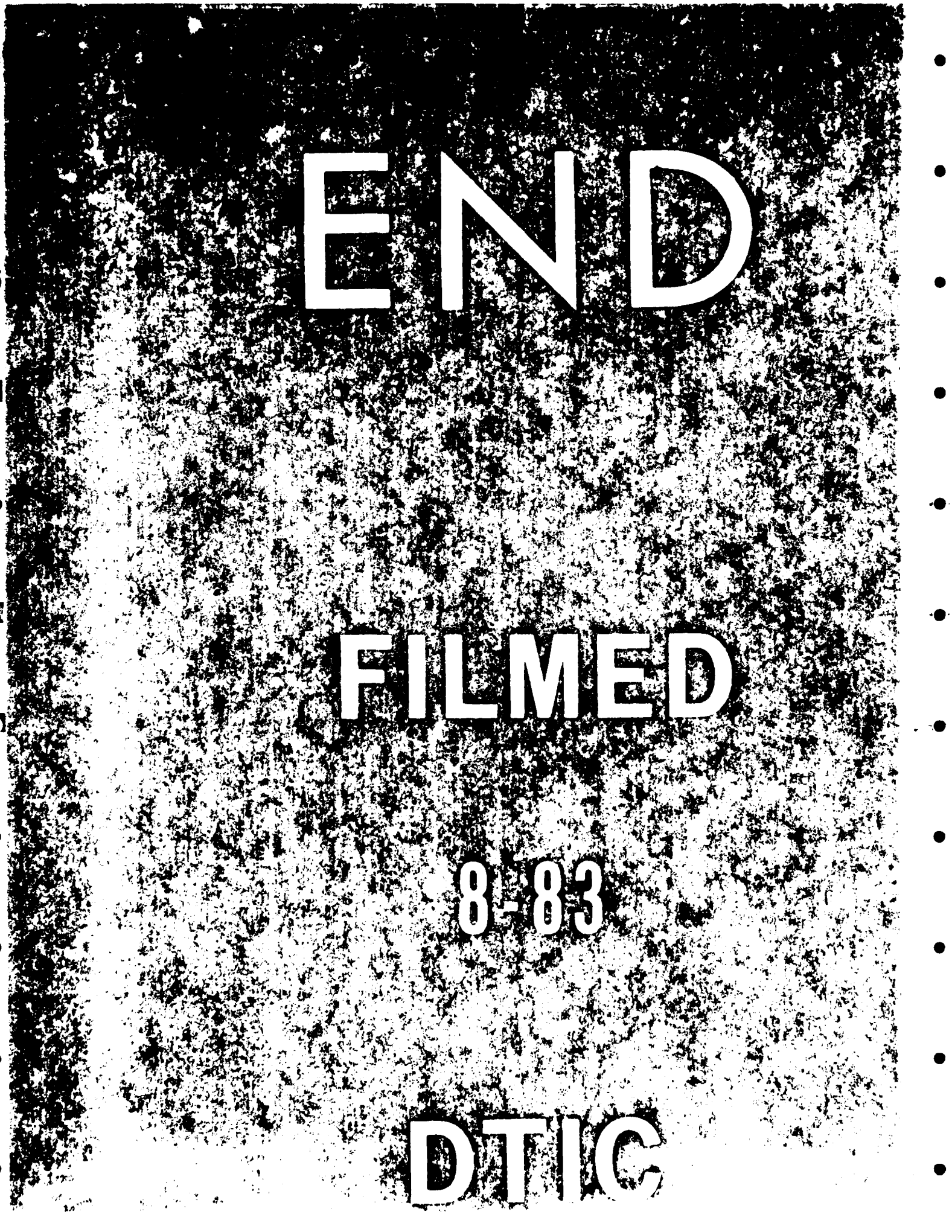
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